JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 103, NO. D6, PAGES 6089–6096, MARCH 27, 1998

On light scattering in random media with large densely packed particles

A. A. Kokhanovsky[1,2]
Earth Observation Research Center, NASDA, Tokyo

Abstract. Simple analytical formulae are presented to calculate the local optical parameters and the spherical albedo r of close-packed media with large particles. The formulae obtained can be used to describe recent experiments on snow and foam optics. It was found that the value of r of weak absorbing semi-infinite layers with large particles depends mostly on two parameters (the effective size of particles and the imaginary part of their refractive index). Another important characteristic is the absorption coefficient of low-level contaminants (soot, dust particles, detergents in foam). This value determines the spherical albedo of snow and foam in the visible. The accuracy of obtained equations is better than 10% at $r > 0.2$.

1. Introduction

The radiative transfer in random media with low concentrations of scatters c (e.g., water and ice clouds, mists, aerosols, hydrosols, interstellar dust) was studied by many authors [*Sobolev*, 1956; *Chandrasekhar*, 1960; *Ambarzumian*, 1960; *van de Hulst*, 1980; *Lenoble*, 1985]. As a result, at present there is no problem to solve the radiative transfer equation (RTE) for a plane-parallel light scattering medium of any optical thickness at $c \ll 1$. A lot of analytical, approximate, and numerical methods of the radiative transfer equation solutions were developed.

In the case of intermediate or high concentrations of particles ($c > 0.1$) the situation is completely different. Generally speaking, the standard radiative transfer equation cannot be used in this case, and one is to apply the T-matrix approach [*Waterman and Truell*, 1961] or different approximate techniques like *Foldy* [1945], *Twersky* [1962], or quasi-crystalline [*Lax*, 1952] approximations. It is a straightforward matter to investigate light scattering and propagation in media with small particles ($\rho < 1$–10, $\rho = ka$, $k = 2\pi/\lambda$, λ is the wavelength, a is the radius of a particle) with these methods. In this case the scattering of radiation is weak. Moreover, different procedures of the replacement of an actual inhomogeneous medium with a uniform one, but with the "effective" refractive index [*Tsang and Kong*, 1983; *Tsang et al.*, 1985], can be used. Such approximations are very important in the microwave region of the electromagnetic spectrum.

For the optical band the dimension of particles in many geophysical media (soils, snow fields, oceanic whitecaps) is very large in comparison with the wavelength, and the scattering of radiation is extremely important. Theoretical methods [*Tsang et al.*, 1985], which appeared to be successful in the microwave region, encounter many obstacles. It is not possible to avoid the difficulties with numerical algorithms of the dense medium theory at large values of the size parameter ka, at least in the nearest future.

Moreover, there is no consensus even on the single light scattering properties of close-packed media with large particles. For example, it follows from experiments [*Ishimaru and Kuga*, 1982; *Ivanov et al.*, 1988] that the ratio of the scattering coefficient σ_{sca} of dense media with large particles to the value of the scattering coefficient of the same medium at $c \ll 1$ increases with the concentration of particles. This seems contrary to the recent experiments [*Gobel*, 1995]. Some authors [e.g., *Wiscombe and Warren*, 1980] advocate that close-packed effects do not take place for large particles. Different opinions about the influence of the cooperative effects on the absorption coefficient σ_{abs} exist as well [*Blevin and Brown*, 1961; *Khairullina*, 1982; *Ivanov et al.*, 1988].

One can see the difficulties that arise in the problem of light scattering and propagation in close-packed media. Thus their solution is to be one of the most important tasks in both theoretical and experimental work in the optics of turbid media in the coming years.

Since there is no exact solution of the problem under discussion, even rough approximations should be applied and experimentally verified [*Hapke*, 1993]. One of them is the use of the effective radiative transport equation (ERTE) to solve the problem [*Hapke*, 1981; *Tsang et al.*, 1990; *Mishchenko*, 1994]. This equation has the same structure as the ordinary equation, but the relation of the phase function $p(\theta)$ (or the scattering matrix), the extinction coefficient, and the single-scattering albedo ω to the optical characteristics of individual particles differs from one for dilute media. It is worth noting that it is possible to use the well-developed methods [*Rozenberg*, 1969; *Ishimaru*, 1978; *Zege et al.*, 1991a; *Stephens*, 1994] of the RTE solution within the framework of this approximation. Some recent experiments and theoretical investigations [*Wiscombe and Warren*, 1980; *Whitlock et al.*, 1982; *Bohren*, 1987; *Zege et al.*, 1991b; *Grenfell*, 1994] show that there are grounds for such an approach (at least, for optically thick weak absorbing media).

The structure of this paper is as follows: In the first part, simple analytical solutions for the phase function, the scattering coefficient, the asymmetry parameter, and the transport mean free path length of close-packed media with large parti-

[1]On leave from the Stepanov Institute of Physics, Minsk, Belarus.
[2]Now at IMVT/UVT, Technical University of Clausthal, Clausthal-Zellerfeld, Germany.

Paper number 97JD03632.
0148-0227/98/97JD-03632$09.00

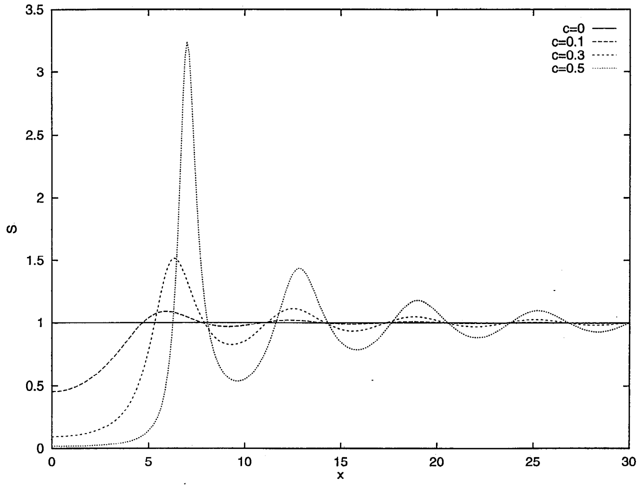

Figure 1. Dependence of the structure factor on parameter $x = 2\theta\rho$ at different values of concentration c.

cles are obtained and analyzed. In the second part, the solution of the RTE for a semi-infinite weak absorbing medium with effective local optical properties obtained is used to describe experiments on snow and foam optics.

2. Single Light Scattering by Densely Packed Large Particles

It is a well-known fact [*Balescu*, 1975; *Rytov*, 1978] that the differential scattering cross section (DSCS) $C_{\text{sca}}(\theta)$ of close-packed media is given by the following equation:

$$C_{\text{sca}}(\theta) = C^0_{\text{sca}}(\theta)S(\theta) \tag{1}$$

where θ is the scattering angle, $C^0_{\text{sca}}(\theta)$ is the DSCS at low concentrations of particles $c \ll 1$. The static structure factor $S(\theta)$ accounts for the correlation effects in dense random media. It follows within the framework of the Ashcroft-Lekner hard-sphere model [*Ashcroft and Lekner*, 1966], based on the Wertheim-Thielle [*Wertheim*, 1963; *Thiele*, 1963] solution of the Percus-Yevick [*Percus and Yevick*, 1958] equation:

$$S(\theta) = \frac{1}{1 - H(\theta)}, \tag{2}$$

where

$$H(\theta) = -\frac{24c\zeta(\theta)}{x^6}, \tag{3}$$

$$\begin{aligned}\zeta(\theta) &= \delta x^3(\sin x - x\cos x) + \beta x^2(2x\sin x \\ &\quad + (2 - x^2)\cos x - 2) \\ &\quad + \Delta((4x^3 - 24x)\sin x - (x^4 - 12x^2 + 24)\cos x + 24),\end{aligned}$$

and $\delta = (1 + 2c)^2/(1 - c)^4$, $\beta = -6c[(1 + c/2)^2]/(1 - c)^4$, $\Delta = \delta c/2$, $x = 2\theta\rho$, $\rho = ka$, $c = N\bar{v}$, $\bar{v}$ is the average volume of particles, N is the number concentration of particles. The structure factor $S(\theta) = C_{\text{sca}}(\theta)/C^0_{\text{sca}}(\theta)$, calculated with (2) and (3) is presented in Figure 1.

Note that $H(0) = 1 - \delta$ and $S(0) = 1/\delta$. Thus it follows

$$C_{\text{sca}}(0) = \frac{(1 - c)^4}{(1 + 2c)^2} C^0_{\text{sca}} \tag{4}$$

and the intensity of light scattering in the forward direction decreases with increasing the concentration of particles. The same effect is in the problem of light scattering by two-sphere clusters [*Mishchenko*, 1996].

Note that simple approximations (2) and (3) provide a good accuracy in the comparison with the numerical solution of the Percus-Yevick equation and Monte Carlo methods [*Ashcroff and Lekner*, 1966]. They have already been used in numerous papers [*Barabanenkov*, 1982; *Saulnier et al.*, 1990; *Mishchenko*, 1992, 1994; *Mishchenko and Macke*, 1997] for studies of close-packed effects in the single-scattering approximation. The value of the DSCS was calculated with the Mie theory in these papers. Here the special case of large particles ($\rho \gg 1$, $2\rho|m - 1| \gg 1$, $m = n - i\chi$ is the refractive index of particles) will be considered. It allows to obtain an analytical solution for the DSCS of a close-packed particulate medium and clarifies the physical mechanism of light scattering by such disperse systems.

In the framework of the geometrical optics approximation (GOA) with account for the diffraction effects it follows [*Shifrin*, 1951; *van de Hulst*, 1981]

$$C^0_{\text{sca}}(\theta) = C^D_{\text{sca}}(\theta) + C^G_{\text{sca}}(\theta), \tag{5}$$

where [*Born and Wolf*, 1959]

$$C^D_{\text{sca}} = 0.25k^2a^4F(z) \tag{6}$$

accounts for the diffraction and

$$C^G_{\text{sca}}(\theta) = 0.25a^2 \sum_{p=0}^{\infty} \frac{\varepsilon_{1p} + \varepsilon_{2p}}{\sin\theta D(\theta)} \sin 2\tau \tag{7}$$

accounts for the geometrical optics scattering of light by spherical particles. Here

$$F(z) = 4J_1^2(z)/z^2, \qquad z = \theta\rho,$$

$$\varepsilon_{jp} = \delta_{0p}R_j + (1 - R_j)^2(1 - \delta_{0p})R_j^{p-1}\exp(-pb\xi),$$

$$\xi = \sqrt{1 - \cos^2\tau^*}, \qquad R_1 = \frac{\tan^2(\tau - \tau^*)}{\tan^2(\tau + \tau^*)},$$

$$R_1 = \frac{\sin^2(\tau - \tau^*)}{\sin^2(\tau + \tau^*)}, \quad \tau^* = \arccos\left(\frac{\cos\tau}{n}\right), \quad D(\theta) = \frac{d\theta}{d\tau},$$

$$b = 4\chi\rho, \qquad \theta = q(2p\tau^* - 2\tau + 2\pi k),$$

δ_{0p} is the Kroneker's symbol, $J_1(z)$ is the Bessel function, τ is the incidence angle. Integer values q, p, k provide the condition $0 \le \theta \le \pi$ for the scattering angle θ. It follows from (6) and (7) that $C^D_{\text{sca}}(\theta) \gg C^G_{\text{sca}}(\theta)$ at $\theta \le \theta_0$, where $\theta_0 \approx 10/\rho$. However [*Ivanov et al.*, 1988; *Balesku*, 1975], the structure factor $S(\theta)$ (2) is almost equal to 1 at $\theta \ge \theta_0(z \ge 20$; see Figure 1). Therefore it follows from (1) and (5) approximately that

$$C_{\text{sca}}(\theta) = C^D_{\text{sca}}(\theta)S(\theta) + C^G_{\text{sca}}(\theta). \tag{8}$$

This result is important and holds for nonspherical particles as well. It follows from (8) that correlation effects change only the diffraction part of the scattered intensity. It is possible to arrive at this conclusion from another point of view. Namely, the diffraction component of the light field is due to the interference of the incident and the scattered waves, and it is partially coherent. The geometrical optical component is the in-

coherent part of the scattering field. Therefore close-packed media phenomena which originate from the interference effects can modify only the first term in (5) and (8).

Results of the computation of the value $\Phi(z) = F(z)S(z)$ (see (6) and (8)) are given in Figure 2 for different values of the concentration c. Note that $\Phi(0) = (1 - c)^4/(1 + 2c)^2$ and $\Phi(0) \to 0$ at $c \to 1$. Thus even small concentrations of particles (10%) produce a significant drop of the value of the phase function in the forward direction [*Lock and Chiu*, 1994]. It is worth pointing out that correlations change the phase function mostly in the region of the forward peak up to $z = 3.83$ and slightly moves the maximum of the scattering intensity to the larger angles.

It is interesting to study the total scattering cross-section C_{sca} in this approach:

$$C_{\text{sca}} = \int_{4\pi} C_{\text{sca}}(\theta)\, d\Omega. \tag{9}$$

From (8) and (9) it follows

$$C_{\text{sca}} = C^D_{\text{sca}} + C^G_{\text{sca}}, \tag{10}$$

where [*Kokhanovsky and Zege*, 1995]

$$C^D_{\text{sca}} = 2\pi a^2 \int_0^\infty \frac{J_1^2(\theta\rho)}{\theta} S(\theta)\, d\theta \tag{11}$$

$$C^G_{\text{sca}} = 0.5\pi a^2 \sum_{j=1}^{2} \int_0^{\pi/2} F_j(\tau) \sin 2\tau\, d\tau. \tag{12}$$

Here $F_j(\tau) = R_j + (1 - R_j)^2 \exp(-b\xi)/(1 - R_j \exp(-b\xi))$. Note that for nonabsorbing particles ($b = 0$) it follows that $F_j(\tau) = 1$ and $C^G_{\text{sca}} = \pi a^2$. There is an approximate solution for

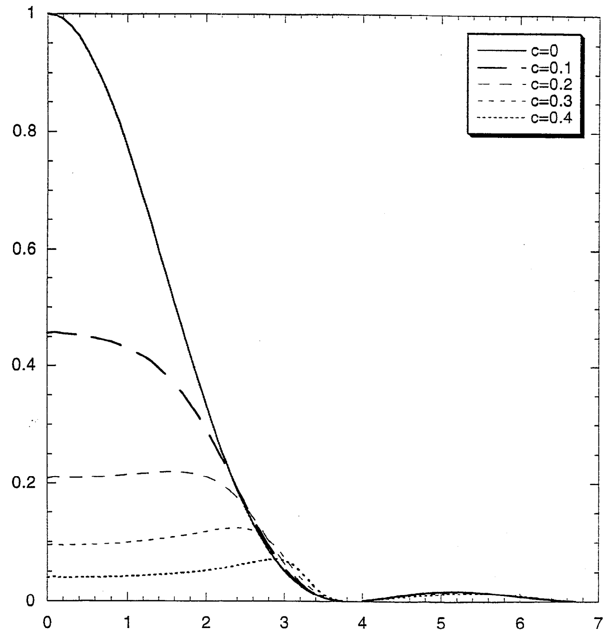

Figure 2. Dependence of function Φ on parameter $z = \theta\rho$ at different values of concentration c.

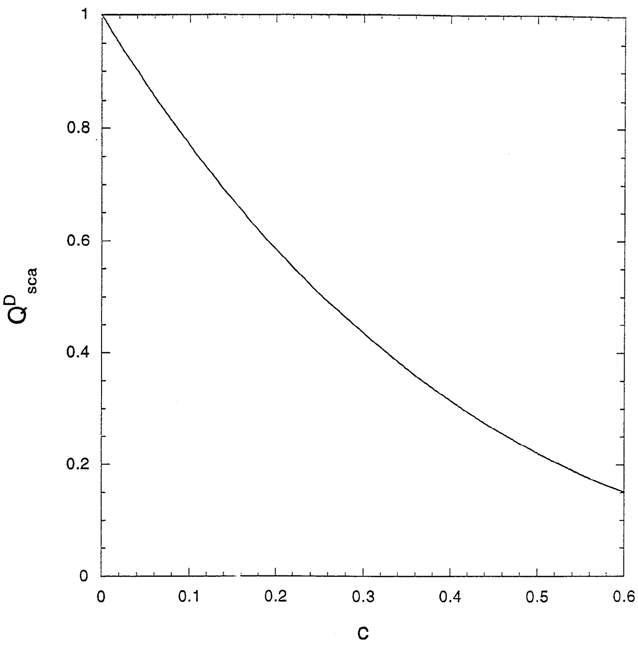

Figure 3. Dependence of Q^D_{sca} on concentration c.

the value of C^G_{sca} at any absorption [*Kokhanovsky and Zege*, 1995] as well:

$$C^G_{\text{sca}} = \pi a^2\{\phi(b) + M(1 - \exp(-b\Delta))^2\}, \tag{13}$$

where

$$\varphi(b) = 2n^2 b^{-2}\{(1 + b\Delta)\exp(-b\Delta) - (1 + b)\exp(-b)\},$$

$$\Delta = \sqrt{1 - n^{-2}}, \qquad M = \frac{1}{2}\int_0^{\pi/2} (R_1 + R_2) \sin 2\tau\, d\tau.$$

To obtain (11), it was taken into account that $C^D_{\text{sca}}(\theta) \to 0$ at $\theta \gg \theta_0$, and therefore it is possible to change the upper limit of the integration from π to ∞. At $c \to 0$, it follows that $S(\theta) \to 1$ and $C^D_{\text{sca}} = \pi a^2$ (see (11)), as it should be at low concentrations.

Results of calculation of the efficiency factor $Q^D_{\text{sca}} = C^D_{\text{sca}}/\pi a^2$ are presented in Figure 3. It is seen that the efficiency factor changes considerably with the concentration of particles. At $c \to 1$, it follows $Q^D_{\text{sca}} \to 0$ and $C_{\text{sca}} \to C^G_{\text{sca}}$. Note that the curve in Figure 3 within the framework of the first coarse approximation can be represented by the following equation: $Q^D_{\text{sca}} = (1 - c)^2$. Therefore the cooperative effects in media with large particles is to decrease the value of the scattering coefficient. This conclusion is supported by *Gobel* [1995], *Hapke* [1981], *Barabanenkov* [1982], *Mishchenko* [1994], and *Saulnier* [1990], but it seems contrary to the results of *Ishimaru and Kuga* [1982] and *Ivanov et al.* [1988]. This discrepancy has to be understood in the future work.

Another important characteristic of the radiative transport in a scattering medium is the phase function $p(\theta)$ and the asymmetry parameter $g = \frac{1}{2}\int_0^\pi p(\theta) \sin\theta \cos\theta\, d\theta$. The phase function is normalized by the following equation:

$$\int_{4\pi} p(\theta)\, d\Omega = 4\pi \tag{14}$$

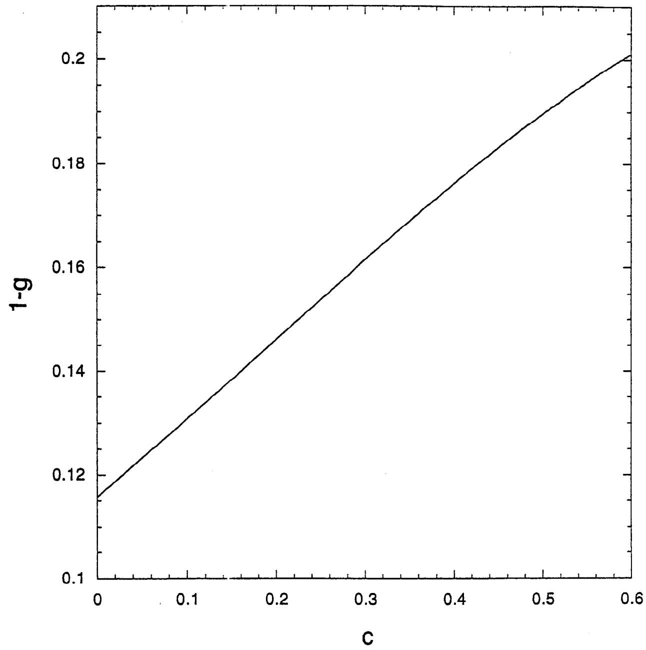

Figure 4. Dependence of 1-g on concentration c.

and it can be obtained from (8) and (10):

$$p(\theta) = \frac{p^D(\theta)C^D_{\text{sca}} + p^G(\theta)C^G_{\text{sca}}}{C^D_{\text{sca}} + C^G_{\text{sca}}}, \tag{15}$$

where

$$p^D(\theta) = \frac{4\pi C^D_{\text{sca}}(\theta)}{C^D_{\text{sca}}}, \tag{16}$$

$$p^G(\theta) = \frac{4\pi C^G_{\text{sca}}(\theta)}{C^G_{\text{sca}}}. \tag{17}$$

For the value of the asymmetry parameter it follows

$$g = \frac{g^D C^D_{\text{sca}} + g^G C^G_{\text{sca}}}{C^D_{\text{sca}} + C^G_{\text{sca}}}, \tag{18}$$

where $g^D \approx 1$ and

$$g^G = \frac{1}{2C^G_{\text{sca}}} \cdot \sum_{j=1}^{2} \int_0^{\pi/2} \frac{K_j(\tau)\sin 2\tau d\tau}{1 - 2R_j \exp(-b\xi)\cos(2\tau^*) + R_j^2 \exp(-2b\xi)}, \tag{19}$$

$$K_j(\tau) = \exp(-b\xi)(1 - R_j)^2 \cos(2(\tau - \tau^*)) + R_j(1 - \exp(-2b\xi))\cos 2\tau + 2R_j^2(\exp(-b\xi) - \cos 2\tau^*)\exp(-b\xi)\cos 2\tau^*. \tag{20}$$

The function $1 - g(c)$ at $n = 1.333$ is presented in Figure 4. It increases approximately half as much as c increases from 0 to 0.6 and can be represented by the following equation: $1 - g = 0.116 + 0.15c$ at $c < 0.6$. Thus correlation effects decrease the asymmetry parameter.

In conclusion of this section the value of the transport mean free path length l_0 for nonabsorbing scatters is to be considered:

$$l_0 = \frac{1}{\sigma_{\text{sca}}(1 - g)}, \tag{21}$$

where $\sigma_{\text{sca}} = NC_{\text{sca}}$, N is the number concentration of particles. The photon transport in the diffuse regime depends on the value of l_0 mostly. For example, it follows [*Zege et al.*, 1991a, b] for the spherical albedo r of a thick nonabsorbing layer that $r = 3L/4l_0[1 + 3L/4l_0]^{-1}$, where L is the geometrical thickness of a medium. Note that the definition of the spherical albedo is the following: $r = 2/\pi \int_0^{2\pi} d\psi \int_0^1 \mu\, d\mu \int_0^1 \mu_0\, d\mu_0 R(\mu, \mu_0, \psi)$. Here $R(\mu, \mu_0, \psi)$ is the reflection function, μ, μ_0 are the cosines of the angles of reflected and incident radiation, ψ is the relative azimuth.

From (18) and (21) it follows

$$l_0 = \frac{1}{NC^G_{\text{sca}}(1 - g^G)}. \tag{22}$$

Note that the value of l_0 does not depend on the partially coherent diffracted part of the light field. This result is important. Indeed, it follows from (22) that one can neglect close-packed effects in diffuse regime for nonabsorbing (or weak absorbing) media with large particles, because of their mutual cancellation. It is the genuine reason of the successful use of the standard RTE in blood [*Dubova et al.*, 1977], foam [*Zege et al.*, 1991a, b], and snow [*Grenfell et al.*, 1994] optics. Of course, it is not true for small ($a \ll \lambda$) particles. However, even in this case the cooperative effects are more important for the scattering cross section than for the transport mean free path length [*Saulnier et al.*, 1990].

3. Spherical Albedo of Semi-infinite Weak Absorbing Media With Large Particles

3.1. Reflection of Light From Snow

From the point of view of light scattering media optics, snow is a collection of densely packed ice grains of different shapes and sizes [*Grenfell et al.*, 1994, Figure 1]. The usual approach for calculating local optical properties of snow media is the Mie theory [*Wiscombe and Warren*, 1980; *Warren*, 1982]. Effects of the nonsphericity and close packing are neglected.

Multiple light scattering in the snowpack is computed by using a multilayer extension of the δ-Eddington approximation [*Wiscombe and Warren*, 1980], two-stream approximation [*Bohren*, 1987], or four-stream [*Grenfell*, 1992] discrete ordinate models. These approaches have already been applied for the interpretation of spherical and plane albedos of snow surfaces [*Wiscombe and Warren*, 1980].

In this paper we will propose a simple model for calculating the spherical albedo of semi-infinite layers of snow, which accounts for the shape of particles. Note that layers of snow with the geometrical thickness L about 5–10 cm can be considered semi-infinite. Indeed, the extinction coefficient σ_{ext} of large particles can be obtained with the following equation [*Kokhanovsky and Macke*, 1997]:

$$\sigma_{\text{ext}} = \frac{1.5c}{a}, \qquad a = \frac{3\bar{v}}{\bar{\Sigma}}, \tag{23}$$

where a is the effective size of grains, $\bar{v}$ and $\bar{\Sigma}$ are their average volume and surface area. At characteristic values of $a = 150$

μm and $c = 0.3$, one obtains that the extinction coefficient is about 30 cm^{-1} and the optical thickness is equal to 150 at $L =$ 5 cm.

Light absorption by ice grains is low in the visible and near infrared. Thus to calculate the spherical albedo of snow, the following analytical solution of the radiative transfer equation at the single-scattering albedo ω, closed to unity, can be used [*Rozenberg*, 1969; *Zege et al.*, 1991a, b]:

$$r = \exp\left(-4y/\sqrt{3}\right), \tag{24}$$

where $y = \sqrt{\sigma_{\text{abs}} l}$, $l = 1/\sigma_{\text{ext}}(1 - g)$. The accuracy of this formula is about 10% at $r > 0.2$. For the weak absorbing media it follows that $\sigma_{\text{ext}} \approx \sigma_{\text{sca}}$ and $l \approx l_0$. Thus the dependence of l on the cooperative effects is weak (see (22)). Such dependence for the absorption coefficient σ_{abs} of media with large particles is weak as well [*Gobel*, 1995]. Indeed, the influence of light rays, passing near a large particle ($ka \gg 1$), on the value of the absorption cross section of a particle is small ($\propto (ka)^{-2/3}$ [*Nussenzveig and Wiscombe*, 1980]). Thus only light incident on the surface of particles is important. It is not effected by the presence of neighbors.

Thus we come to the conclusion that the simple equation (24) can be used to estimate the reflection function of semi-infinite close-packed media with large weak absorbing particles, and "effective" optical parameters coincide with "real" ones, which can be calculated with the geometrical optics approach (or the Mie theory). This result is important from the point of view of possible spectroscopic applications (the spectroscopy of snow and foam) and is to be verified in laboratory and field experiments.

Within the framework of the geometrical optics approach for large weak absorbing particles of any shape it follows [*Kokhanovsky and Zege*, 1995, 1997; *Kokhanovsky and Macke*, 1997]:

$$\sigma_{\text{abs}} = f(n, s)c\alpha, \tag{25}$$

where $\alpha = 4\pi\chi/\lambda$. The function $f(n, s)$ and the asymmetry parameter g depend on the value of the real part of the refractive index of particles n and their shape (via the vector-parameter s). Note that they almost do not depend on the size of particles in the visible and near infrared. The value of n for water and ice depends only slightly on the wavelength λ in the visible and near infrared. Thus it follows from (23) to (25) that

$$r = \exp\left(-A\sqrt{\alpha(\lambda)a}\right), \tag{26}$$

where the parameter $A = 4/3\sqrt{2f(n, s)/(1 - g(n, s))}$ depends on the shape and the real part of the refractive index, but it practically does not depend on the wavelength and size. One can see that indeed, it is possible to use the approximation of spherical particles to investigate the spectral spherical albedo of media with large weak absorbing nonspherical scatters [*Wiscombe and Warren*, 1980]. The radius of the equivalent spherical particles $a = \gamma^2 a_n$, where a_n is the effective size of nonspherical particles, and γ is the ratio of values of A for nonspherical particles to this value for spheres.

Calculations show [*Kokhanovsky and Macke*, 1997] that both the values of $1 - g$ and f increase with the nonspherity of particles, but the value of $A \propto \sqrt{f(n, s)/1 - g(n, s)}$ does not change considerably ($A \approx 5 \div 7$). We assume in this paper that $A = 6$ for snow media (as for water clouds at $n = 1.3$) and

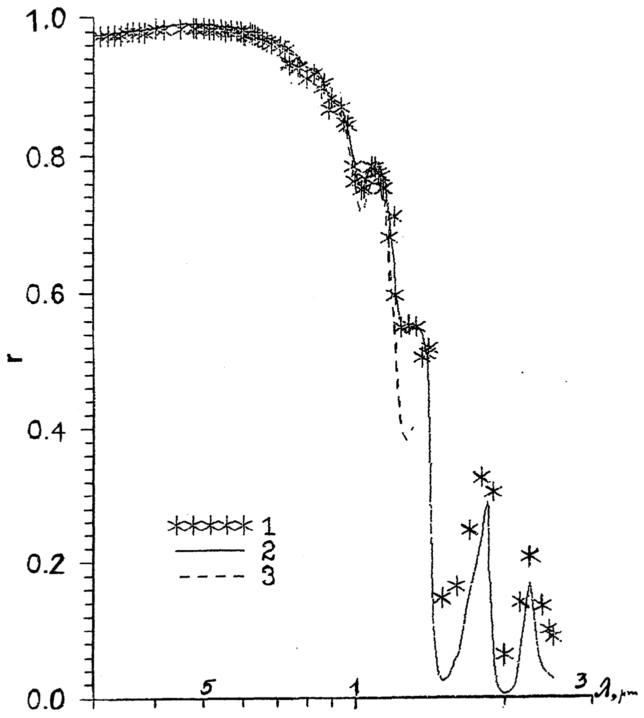

Figure 5. Dependence of the spherical albedo of snow r on wavelength λ(1, experiment [*Grenfell et al.*, 1994]; 2, equation (26) at $A = 6$, $a = 80$ μm; 3, asymptotical equation $r = 1 - 6\sqrt{\alpha(\lambda)a}$).

$$r = \exp\left(-6\sqrt{\alpha(\lambda)a}\right). \tag{27}$$

To study the accuracy of (27) for the spherical albedo of snow, we compared the results of the calculation with (27) and the results of field experiments on light reflectance from snow in the Antarctic [*Grenfell et al.*, 1994]. The results of the calculations are presented in Figure 5 at $a = 80$ μm, which is close to the mean size of grains in the upper snow layer, simultaneously measured during the experiment. One can see that the accuracy of the simple equation (27) is high, especially at $r >$ 0.2, where the general equation (24) is valid. Note that at $\lambda <$ 1.2 μm, an asymptotical relation $r = 1 - 6\sqrt{\alpha(\lambda)a}$ [*Bohren and Barkstrom*, 1974], which follows from (27) at small values of $\alpha(\lambda)a$, can be used (see Figure 5).

Measurements by *Grenfell et al.* [1994] were carried out in the Antarctic. This is the reason of the high values of the spherical albedo of snow at the visible wavelengths in Figure 5. Snow in other continents contains a lot of impurities [*Warren and Wiscombe*, 1980]. To describe the effects of pollutants, (25) and (27) should be modified. Namely, instead of (25) one obtains

$$\sigma_{\text{abs}} = fc\alpha + \sum_{i=1}^{M} f_i c_i \alpha_i \tag{28}$$

where α_i, c_i, f_i are the absorption coefficient, the concentration, and the shape-dependent parameter of i-impurity (soot, heavy metals, dust, etc.). Neglecting the scattering of light by contaminants, it is possible to obtain from (23), (24), and (28)

$$r = \exp\left[-A\sqrt{(\alpha(\lambda) + \gamma(\lambda))a}\right], \tag{29}$$

where

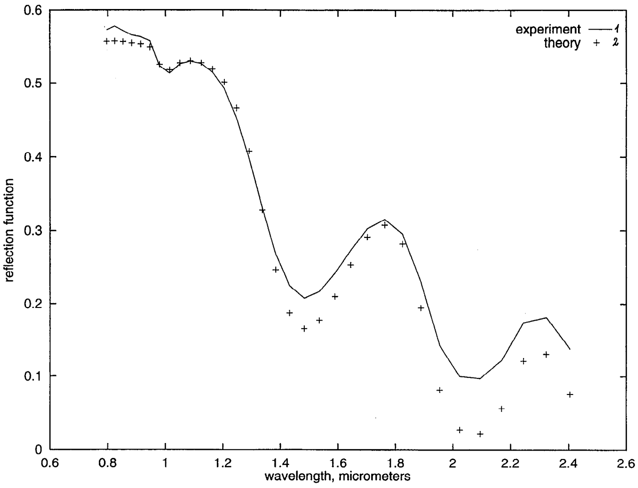

Figure 6. Dependence of the spherical albedo r of foam on the wavelength λ (1, experiment for directional reflectances [*Whitlock et al.*, 1982]; 2, equation (29) at $A = 1.5$, $a = 1000$ μm, $\gamma = 0.00015\ \mu\text{m}^{-1}$).

$$\gamma(\lambda) = \frac{1}{cf}\sum_{i=1}^{M} f_i c_i \alpha_i. \tag{30}$$

The second term in (29) can be neglected at $\lambda > 1\ \mu$m [*Warren*, 1982].

3.2. Reflection of Light From Whitecaps

Another example of light-scattering close-packed media with large particles is oceanic whitecaps. Their radiative properties were studied theoretically by *Stabeno and Monahan* [1986] and *Zege et al.* [1991b]. It was shown in the last paper that (24) can be applied for calculations of the spectral reflectance of foams.

In the case of foam the extinction coefficient $\sigma_{\text{ext}} = 1.5ca^{-1}$ as for snow grains, but the absorption takes place not in particles (air bubbles) but between them in liquid films and [*Kokhanovsky*, 1989] $\sigma_{\text{abs}} = f(n, s)(1 - c)\alpha$. Thus it follows from (23) and (26) that $A = 4/3\ \sqrt{2f(n,s)(1-c)/(1-g(n,s))c}$ for foams. This value depends on the concentration of bubbles c. The dependence of A on the wavelength is weak in the visible and near-infrared regions of the electromagnetic spectrum. The value of A for spherical bubbles is about 0.6 [*Kokhanovsky*, 1989] at $c = 0.99$. It is worth pointing out that the spectral dependence of the spherical albedo of foam with $A = 0.6$ and $a = 1000\ \mu$m coincides with the spectral dependence of this function for water clouds at $a = 10\ \mu$m ($A = 6$) within the framework of this approximation.

Note that foam cannot be produced from pure water. Thus there are different low-level contaminants in foam (like in snow). They determine the spectral behavior of foam in the visible. This is the reason of different spectral dependencies of the spherical albedo of foam, obtained by different authors [*Whitlock et al.*, 1982; *Frouin et al.*, 1997; *Moore et al.*, 1997]. The spherical albedo of foam with account for contaminates can be obtained from (29), where

$$A = \frac{4}{3}\sqrt{\frac{2f(n, s)(1 - c)}{(1 - g(n, s))c}}, \tag{31}$$

$$\gamma(\lambda) = \frac{1}{(1 - c)f}\sum_{i=1}^{M} f_i c_i \alpha_i. \tag{32}$$

Let us compare calculations of the spherical albedo r with (29), (31), and (32) with laboratory experiments on foam reflectance, performed by *Whitlock et al.* [1982]. The spectral behavior of the foam reflectance was almost neutral and equal approximately 0.6 in the visible at the experiment. Thus one can obtain from (29), (31), and (32) that $\gamma = 0.00015\ \mu\text{m}^{-1}$ in this experiment. The results of calculations of $r(\lambda)$ for foam at $A = 1.5 (c = 0.94)$, $a = 1000\ \mu$m, and $\gamma = 0.00015\ \mu\text{m}^{-1}$ using (29) are presented in Figure 6. One can see that our simple equation (29) can be used for the interpretation of the foam reflectance spectra at $r > 0.2$.

Note that (29) can be used to find the spectral dependence of absorption coefficients of contaminants from the reflectance measurements. Indeed, from (29) it follows in the visible that

$$\Gamma(\lambda) = \frac{\ln^2 r(\lambda)}{\ln^2 r(0.55\ \mu\text{m})}, \tag{33}$$

where $\Gamma(\lambda) = \gamma(\lambda)/\gamma(0.55\ \mu\text{m})$ at $\gamma(\lambda) \gg \alpha(\lambda)$. This equation is important for the spectral analysis of optically thick dispersed media with large weak absorbing particles of any shape and concentration.

It should be pointed out that measurements of foam reflectances in the paper by *Whiltock et al.* [1982] are actually directional measurements and not spherical albedos. The reflection function of the semi-infinite strongly scattering weak absorbing layer can be calculated with the following approximate formula [*Zege et al.*, 1991a]:

$$R(\mu, \mu_0, \psi) = R_0(\mu, \mu_0, \psi)\exp\left\{\frac{-4ys(\mu)s(\mu_0)}{\sqrt{3}R_0(\mu, \mu_0, \psi)}\right\}, \tag{34}$$

where $s(x) = 3/7(1 + 2x)$, $R_0(\mu, \mu_0, \psi)$ is the reflection function of the semi-infinite nonabsorbing medium with the same phase function as the absorbing medium under consideration. The value of $R_0(\mu, \mu_0, \psi)$ is close to 1 for most of the angles, and it follows from (24), (26), and (34) approximately that

$$R(\mu, \mu_0, \psi) = \exp\left(-A\sqrt{\alpha(\lambda)a^*}\right), \tag{35}$$

where $a^* = as^2(\mu)s^2(\mu_0)$. Unfortunately, values of the incident and observation angles were not specified by *Whiltock et al.* [1982]. Thus we assumed in our calculations in Figure 6 that $s^2(\mu)s^2(\mu_0) = 1$. Equations (26) and (35) coincide under this assumption.

4. Conclusions

Generally speaking, there are two types of cooperative effects in dense media, namely, (1) the change of the effective field in a scattering medium and (2) interference effects.

For small particles ($a \ll \lambda$), effects 1 are the most important, and effects 2 can be neglected because of very low light scattering in such media. The main effect is the change of the effective field and the dielectric permittivity.

On the other hand, for large particles ($a \gg \lambda$), effects 2 are more important, but they take place only in the very narrow region of the scattering angles near the forward direction. Interference effects for large particles reduce the scattering coefficient, the asymmetry parameter, and the extinction coef-

ficient of media with large particles considerably, but they do not change the absorption coefficient and the transport mean free path length, which are responsible for the optical properties of light scattering media at the diffuse regime (optically thick weak absorbing layers). Thus it is possible to apply the standard radiative transfer equation to study the light reflectance from dense media with large particles. It is supported by Figures 5 and 6, where the comparisons of theoretical (see (26)) and experimental results for the spherical albedo of snow and foam are given.

Note that (24) can be generalized in the case of finite inhomogeneous layers with the underlying surface [*Zege et al.*, 1991a, b]. The finite depth and a multilayer structure of snow and foam can be fully taken into account in the framework of this approach as well. This will be the subject of our next paper.

At $a \propto \lambda$, effects 1 and 2 are both equally important, and exact methods of the dense media transport theory [*Tsang et al.*, 1985] should be used.

This paper supports experimental results of *Whiltock et al.* [1982], *Frouin et al.* [1997], and *Moore et al.* [1997] that the wavelength dependence of absorption in liquid water strongly impacts measured reflectance of foam. Foam reflectance is not independent of wavelength as has been assumed in some previous studies [*Koepke and Quenzel*, 1981].

Note that recent field experiments, carried out by *Frouin et al.* [1997], show that foam reflectance depends on the wavelength in the visible region of the electromagnetic spectrum (contrary to the laboratory measurements performed by *Whitlock et al.* [1982]). From our point of view this dependence is mostly due to low-level contaminants, always presented in seawater. The chemical composition of these contaminants varies with the location (open ocean, coastal areas, etc.) and so does the reflectance spectrum of the whitecaps in the visible.

Acknowledgments. This work was supported by the Science and Technology Agency of Japan. The author wishes to thank Y. Barabanenkov, A. Ishimaru, L. Tsang, and E. Zege for useful discussions. He is also grateful to the anonymous reviewers for the important advices.

References

Ailawadi, N. K., Generalization of the Ashcroft-Lekner hard-sphere model for the structure factor, *Phys. Rev.*, *A7*, 2200–2203, 1973.

Ambarzumian, V. A., *Scientific Papers*, vol. 1, 428 pp., Akad. Nauk SSSR, Yerevan, 1960.

Ashcroft, N. W., and J. Lekner, Structure and resistivity of liquid metals, *Phys. Res.*, *145*, 83–90, 1966.

Balescu, R., *Equilibrium and Nonequilibrium Statistical Mechanics*, John Wiley, New York, 1975.

Barabanenkov, Y. N., On relative increasing of the extinction length due to correlation of the weak scatterers, *Izv. Akad. Nauk SSSR, Fiz. Atmos. Okean.*, *18*, 720–726, 1982.

Blevin, W. R., and W. J. Brown, Effect of particle separation on the reflectance of semi-infinite diffusers, *J. Opt. Soc. Am.*, *51*, 129–134, 1961.

Bohren, C. F., Multiple scattering of light and some of its observable consequences, *Am. J. Phys.*, *55*, 524–533, 1987.

Bohren, C. F., and B. R. Barkstrom, Theory of optical properties of snow, *J. Geophys. Res.*, *79*, 4527–4535, 1974.

Born, M., and E. Wolf, *Principles of Optics*., Pergamon, Tarrytown, N. Y., 1959.

Chandrasekhar, S., *Radiative Transfer, 393 pp*., Dover, Mineola, N. Y., 1960.

Dubova, G. S., et al., Determination of the absorption spectrum of a hemoglobin by light scattering methods, *J. Appl. Spektr.*, *27*, 871–878, 1977.

Foldy, L. L., The multiple scattering of waves, *Phys. Rev.*, *67*, 107–119, 1945.

Frouin, R., M. Schwinling, and P.-Y. Deschamps, Spectral reflectance of sea foam in the visible and near-infrared: In situ measurement and remote sensing applications, *J. Geophys. Res.*, *102*, 14,361–14,371, 1997.

Gobel, G., Dependent scattering effects in latex-sphere suspensions and scattering powders, *Waves Random Media*, *5*, 413–426, 1995.

Grenfell, T. C., et al., Reflection of solar radiation by the Antarctic snow surface at ultraviolet, visible, and near-infrared wavelengths, *J. Geophys. Res.*, *d99*, 18,669–18,684, 1994.

Grenfell, T. C., A radiative transfer model for sea ice with vertical structure variations, *J. Geophys. Res.*, *96*, 16,991–17,001, 1991.

Hapke, B., Bidirectional reflectance spectroscopy, 1, Theory, *J. Geophys. Res.*, *86*, 3039–3054, 1981.

Hapke, B., *Theory of the Reflectance and Emmitance Spectroscopy*, 455 pp., Cambridge Univ. Press, New York, 1993.

Ishimaru, A., *Wave Propagation and Scattering in Random Media*, vols. 1, 2, 572 pp., Academic, San Diego, Calif., 1978.

Ishimaru, A., and Y. Kuga, Attenuation constant of a coherent field in a dense distribution of particles, *J. Opt. Soc. Am.*, *72*, 1317–1320, 1982.

Ivanov, A. P., et al., *Light Propagation in Close Packed Media*, 120 pp., Nauka and Tekh., Minsk, 1988.

Khairullina, A. Y. Studies of cells by light scattering, in *Light Propagation in a Dispersed Medium*, edited by A. P. Ivanov, pp. 275–292, Nauka and Tekh., Minsk, 1982.

Koepke, P., and H. Quenzel, Turbidity of the atmosphere determined from satellite: Calculation of optimum wavelength, *J. Geophys. Res.*, *86*, 9801–9805, 1981.

Kokhanovsky, A. A., Integral scattering characteristics of large spherical particles with the refractive index below unit, *Opt. Spektr.*, *67*, 165–169, 1989.

Kokhanovsky, A. A., and A. Macke, Integral light scattering and absorption characteristics of large nonspherical particles, *Appl. Opt.*, *36*, 8785–8790, 1997.

Kokhanovsky, A. A., and E. P. Zege, Local optical parameters of spherical polydispersions: Simple approximations, *Appl. Opt.*, *34*, 5513–5519, 1995.

Kokhanovsky, A. A., and E. P. Zege, Optical properties of aerosol particles: A review of approximate analytical solutions, *J. Aerosol Sci.*, *28*, 1–21, 1997.

Lax, M., Multiple scattering of waves, II, The effective field in dense systems, *Phys. Rev.*, *85*, 261–269, 1952.

Lenoble, J. (Ed.), *Radiative Transfer in Scattering and Absorbing Atmospheres: Standard Computational Procedures*, 299 pp., A Deepak, Hampton, Va., 1985.

Lock, J. A., and C. L. Chiu, Correlated light scattering by a dense distribution of condensation droplets on a window pane, *Appl. Opt.*, *33*, 4663–4671, 1994.

Mishchenko, M. I., The angular width of the coherent back scatter opposition effect: An application to icy outer planet, *Astrophys. Space Sci.*, *194*, 327–333, 1992.

Mishchenko, M. I., Asymmetry parameters of the phase function for densely packed scattering grains, *J. Quant. Spectrosc. Radiat Transfer*, *52*, 95–110, 1994.

Mishchenko, M. I., Coherent backscattering by two-sphere clasters, *Opt. Lett.*, *21*, 623–625, 1996.

Mishchenko, M. I., and A. Macke, Asymmetry parameters of the phase function for isolated and densely packed spherical particles with multiple internal inclusions in the geometric optics limit, *J. Quant. Spectr. Radiat. Transfer*, *57*, 767–794, 1997.

Moore, K. D., K. J. Voss, and H. G. Gordon, Whitecaps: Spectral reflectance in the open ocean and their contribution to water-living radiance, *Proc. SPIE Int. Soc. Opt. Eng.*, *2963*, 246–251, 1997.

Nussenzweig, H. M., and W. J. Wiscombe, Efficiency factors in Mie scattering, *Phys. Rev. Lett.*, *45*, 1490–1494, 1980.

Percus, J. K., and G. J. Yevick, Analysis of classical statistical mechanics by means of collective coordinates, *Phys. Rev.*, *110*, 1–13, 1958.

Rozenberg, G. V., Absorption spectroscopy of dispersed materials, *Sov. Phys. Usp.*, *2*, 666–698, 1969.

Rytov, S. M., et al., *Introduction to Statystical Radiophysics*, vol. 2, Nauka, Moscow, 1978.

Saulnier, P. M., et al., Scatterer correlation effects on photon transport in dense random media, *Phys. Rev.*, *B42*, 2621–2623, 1990.

Shifrin, K. S., *Scattering of Light in a Turbid Media*, Gostekhteorizdat, Moscow, 1951.

Sobolev, V. V., *Radiative Transfer in the Stars and Planets' Atmospheres*, 391 pp., Gostekhizdat, Moscow, 1956.

Stabeno, P. J., and E. C. Monahan, The influence of whitecaps on the albedo of the sea surface, in *Oceanic Whitecaps*, edited by E. C. Monahan and G. M. Niocaill, pp. 261–266, D. Reidel, Norwell, Mass., 1986.

Stephens, G. L., *Remote Sensing of Lower Atmosphere*, 523 pp., Oxford Univ. Press, New York, 1994.

Thiele, E., Equation of state for hard spheres, *J. Phys. Chem.*, *39*, 474–479, 1963.

Tsang, L., and J. A. Kong, Scattering of electromagnetic waves from a half space of densely distributed dielectric scatterers, *Radio Sci.*, *18*, 1260–1272, 1983.

Tsang, L., et al., *Theory of Microwave Remote Sensing*, Wiley-Interscience, New York, 1985.

Tsang, L., et al., *Progress in Electromagnetic Research*, vol. 3, edited by J. A. Kong, 75 pp., Elsevier, New York, 1990.

Twersky, V., On scattering of waves by random distributions, I, Free space scatterer formulation, *J. Math. Phys.*, *3*, 700–715, 1962.

van de Hulst, H. C., *Multiple Light Scattering* (Tables, formulas, and applications), 739 pp., Academic, San Diego, Calif., 1980.

van de Hulst, H. C., *Light Scattering by Small Particles*, 470 pp., Dover, Mineola, N. Y., 1981.

Warren, S. G., Optical properties of snow, *Rev. Geophys.*, *20*, 67–89, 1982.

Warren, S. G., and W. J. Wiscombe, A model of the spectral albedo of snow, II, Snow, containing atmospheric aerosols, *J. Atmos. Sci.*, *37*, 2734–2745, 1980.

Waterman, P. C., and R. Truell, Multiple scattering of waves, *J. Math. Phys.*, *2*, 512–537, 1961.

Wertheim, M. S., Exact solution of the Percus-Yevick integral equation for hard spheres, *Phys. Rev. Lett.*, *10*, 321–323, 1963.

Whitlock, C. H., D. S. Bartlett, and E. A. Curganus, Sea foam reflectance and influence on optimum wavelength for remote sensing of ocean aerosols, *Geophys. Res. Lett.*, *9*, 719–722, 1982.

Wiscombe, W. J., and S. G. Warren, A model for the spectral albedo of snow, I, Pure snow, *J. Atmos. Sci.*, *37*, 2712–2733, 1980.

Zege, E. P., et al., *Image Transfer Through a Scattering Media*, Springer-Verlag, New York, 1991a.

Zege, E. P., et al., Phenomenological model of optical properties of close-packed strong scattering media and its application to foam optics, *Opt. Spektr.*, *71*, 835–841, 1991b.

A. A. Kokhanovsky, IMVT/UVT, Technical University of Clausthal, 19 Leibnizstrasse, Clausthal-Zellerfeld 38678 Germany. (e-mail: alex@eorc.nasda.go.jp)

(Received October 28, 1996; revised December 2, 1997; accepted December 2, 1997.)